

Tungsten Fibre Reinforced Zr-based Bulk Metallic Glass Composites

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A Zr-based bulk metallic glass (BMG) alloy with the composition $(\text{Zr}_{55}\text{Al}_{10}\text{Ni}_5\text{Cu}_{30})_{98.5}\text{Si}_{1.5}$ was used as the base material to form BMG composites. Tungsten fiber reinforced BMG composites were successfully fabricated by pressure metal infiltration technique, with the volume fraction of the tungsten fiber ranging from 10% to 70%. Microstructure and mechanical properties of the BMG composites were investigated. Tungsten reinforcement significantly increased the material's ductility by changing the compressive failure mode from single shear band propagation to multiple shear bands propagation, and transferring stress from matrix to tungsten fibers.

Keywords: Bulk metallic glasses; Bulk metallic glass composites; Amorphous metals; Zr-based BMG; Tungsten fibre reinforced BMG; shear band; ductility.

Introduction

The discovery of bulk metallic glasses (BMGs) has created a new and exciting class of materials with unique physical and mechanical properties. BMGs exhibit very high strength, specific strength and elastic strain, along with unusual combinations of other engineering properties, which make them very attractive for structural applications [1-3]. Among all BMGs, multicomponent alloys based on Zr have the best glass-forming ability (GFA). They can be readily produced into parts with size of several centimeters by conventional casting technique [4].

The main technical barrier for BMGs to be widely used as structural materials is their limited ductility [5]. All the BMGs developed so far exhibit none or very limited plastic deformation after yielding. The material form localized shear bands, leading to catastrophic shear failure without plastic strain. In contrast to BMGs, crystalline metals usually exhibit substantial plastic deformation before failure, which is critical in terms of reliability for industrial and structural applications. Therefore in the last few years there are increasing attempts to incorporate crystalline metals into BMGs to form BMG matrix composites [6,7].

Johnson's group has investigated BMG composites of Be-containing Vit1 ($\text{Zr}_{41.25}\text{Ti}_{13.75}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$) reinforced with continuous tungsten and stainless steel fibers [4]. Compressive strain to failure was increased by over 900% compared to the unreinforced BMG materials. In this study, we fabricate tungsten reinforced BMG composites using Be-free Zr-based alloy. Microstructure and mechanical properties of the BMG composites are investigated. Mechanisms for the improvement of mechanical properties are also discussed.

Experimental

$(\text{Zr}_{55}\text{Al}_{10}\text{Ni}_5\text{Cu}_{30})_{98.5}\text{Si}_1$ was chosen as the matrix due to its strong glass forming ability [8]. BMG ingots were made by melting a mixture of pure Zr, Al, Ni, Cu and Si elements (the purity of all elements is above 99.8%) in an induction furnace under Ar atmosphere. Tungsten fiber with a nominal diameter of 250 μm was straightened and cut to 80 mm in length. The tungsten fibers were cleaned in an ultrasonic bath of acetone, followed by ethanol. Composite samples were fabricated with tungsten fiber volume fractions of 10, 20, 30, 40, 50, 60 and 70%. Samples were cast in a resistant furnace by melting the matrix ingot in an evacuated quartz

tube packed with the tungsten fibers, followed by pressure infiltration. After 20 minutes of pressurization the tubes were quenched in a supersaturated brine solution. The cast samples were 6 mm in diameter and 90 mm in length.

X-ray diffraction patterns of a cross sectional slice of each composite were taken using a RIGAKU D/max diffractometer with Cu K α radiation. For compression testing, the castings were first centerless ground and then cut to a length of 12 mm. Compression tests were conducted in an MTS-type axial-torsional load frame. The strain rate was 3.0×10^{-4} /s. Microstructure and fracture surface of each BMG composite were examined using a JEOL JSM6301F scanning electron microscope (SEM).

Results and Discussion

The physical properties of BMG matrix and tungsten fiber in the composites are shown in Table 1 [4]. Figure 1 shows X-ray diffraction patterns of the the BMG composites reinforced with 0%, 10% and 30% tungsten respectively. The monolithic BMG only exhibits a characteristic broad diffraction of metallic glass with no evidence of any crystalline Bragg peaks, indicating the formation of a single amorphous phase. While the patterns of the composites show diffraction peaks from tungsten fiber superimposed on the broad halo diffraction of the amorphous phase. No other crystalline phases are detected within the sensitivity limit of the XRD, which suggests that the matrix is still amorphous after melt infiltration and quenching. Because of its high melting temperature, the microstructure of the tungsten is unaffected during the melt infiltration process.

During the melt infiltration process, the most critical parameter is the infiltration temperature. Figure 2 shows the microstructures of the interface between BMG matrix and tungsten reinforcement in the composites processed at

Table 1.—Physical properties of BMG matrix and tungsten fiber [4].

Young's modulus, GPa	87	410
Shear modulus, GPa	32.1	160.5
Poisson's ratio	0.37	0.28
Ultimate strength, MPa	1830	2100
Tensile yield strength	1830	1650
Tensile strain to failure, %	2	1.9
Hardness (Vickers), kg/mm ²	520	650
Thermal expansion coefficient, $10^{-6}/^{\circ}\text{C}$	8.5	4.5
Density, g/cm ⁻³	6.6	19.3

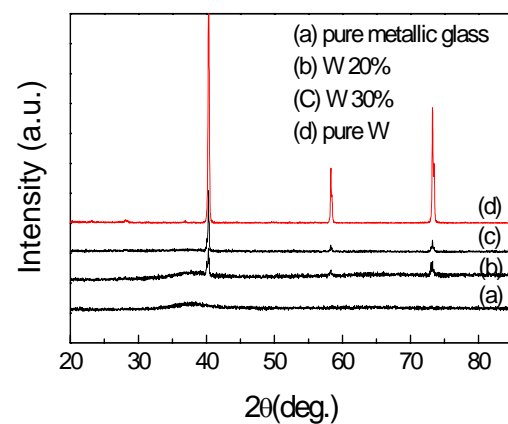


Figure 1.—X-ray diffraction patterns of the BMG composites. The nominal volume fraction of the tungsten reinforcement are (a) 0%; (b) 20% and (c) 30%. The pattern from pure tungsten is also included as reference (d).

different temperatures. At 930°C, sharp W/BMG interface is obtained with no crystalline phase in the matrix (Figure 2a). With the increase of the melt infiltration temperature, small crystals are formed adjacent to the tungsten fibers, as shown in Figure 2b. This crystalline phase could not be detected by X-ray diffraction, however, electron probe analysis identifies it is Zr₂Cu. At a higher temperature 1030°C, besides of the crystallization, we also observed the damage of tungsten fibers (Figure 2c). Figure 2d shows the microstructure of the uniaxially tungsten

Properties	Zr BMG	Tungsten
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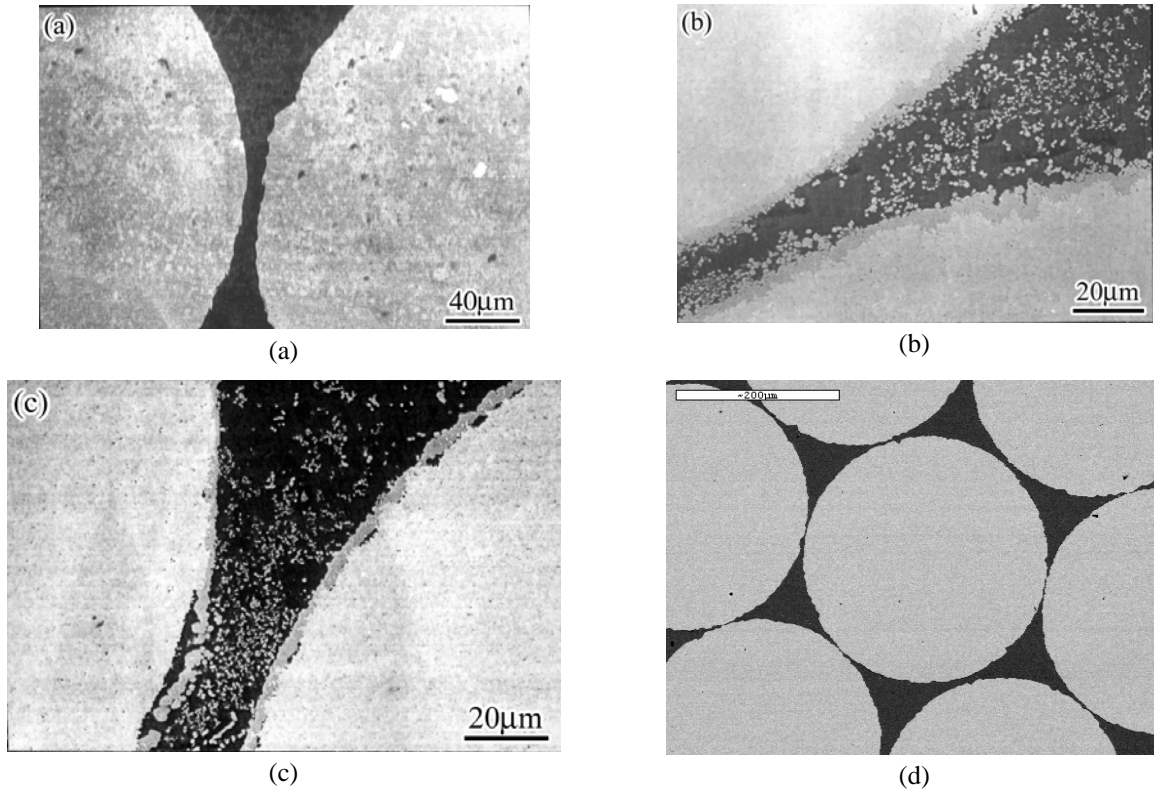


Figure 2.—SEM micrographs of the interface between BMG matrix and tungsten reinforcement in the composites processed at temperature of (a) 930°C; (b) 980°C and (c) 1030°C respectively. (d) is the cross section microstructure of the uniaxially tungsten reinforced BMG composite with 70% W fiber.

reinforced BMG composite. A uniform and homogenous structure is obtained.

Quasi-static compression tests were performed on the BMG composites with tungsten fiber volume fraction of 0, 10, 28, 40, 60 and 70% respectively. At least three samples of each V_f were tested. The stress-strain curves of the materials are shown in Figure 3. The monolithic BMG shows no plastic deformation. It fractured immediately after yielding. The tungsten reinforced BMG composite with over 28% W fiber show elastic-perfectly-plastic stress-strain behavior, with strains of up to 13%.

Figure 4 shows the compressed samples of reinforced and unreinforced BMGs. In the monolithic BMG sample, the material failed by shear mode. The plastic deformation is confined to the shear band region. The compressive fracture took place along the maximum shear plane, which is inclined by about 45° to the

direction of applied load. It was often observed that material failed on one dominant shear band

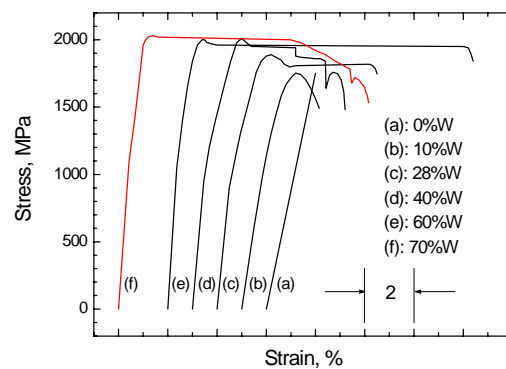


Figure 3.—Quasi-static compressive stress-strain curves of BMG composites with different level tungsten volume fraction.

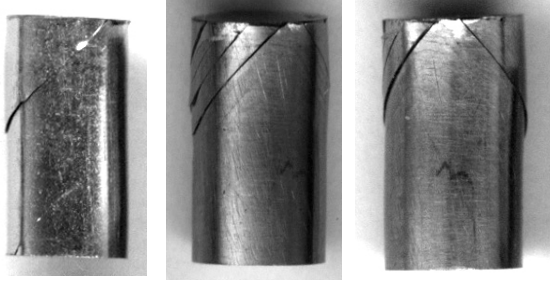


Figure 4.—Shear band formation during compressive test: (a) single shear band in unreinforced BMG; (b) multiple shear bands in BMG composite with 40% tungsten fiber, and (c) formation of two cross-shear bands. The diameter of the test samples is 6 mm.

with very little plastic strain, as shown in Figure 4a. In BMG composite materials, the tungsten reinforcement acts as a “crack stopper” by adding impediments to shear band propagation and promoting multiple shear bands. Figure 4b shows the multiple shear bands being formed in

the material with 40% tungsten fiber. When the fiber volume fraction is high, the matrix is highly constrained, and only small slip displacement can occur, therefore stress concentration builds up at the tip of a propagating shear band. When the shear band meets an obstacle, this stress concentration triggers the formation of another shear band, typically at an angle of 90° to the first shear band, as shown in Figure 4c.

Figure 5 shows the SEM micrographs of the BMG composite reinforced with 60% tungsten fiber, revealing the shear band formation and propagation at different stages of compression test. In Figure 5a, the test was interrupted just after the yielding point. It reveals that after yielding multiple shear bands are formed in glass. The formation of the multiple shear bands is initiated by the reinforcement fibers, which

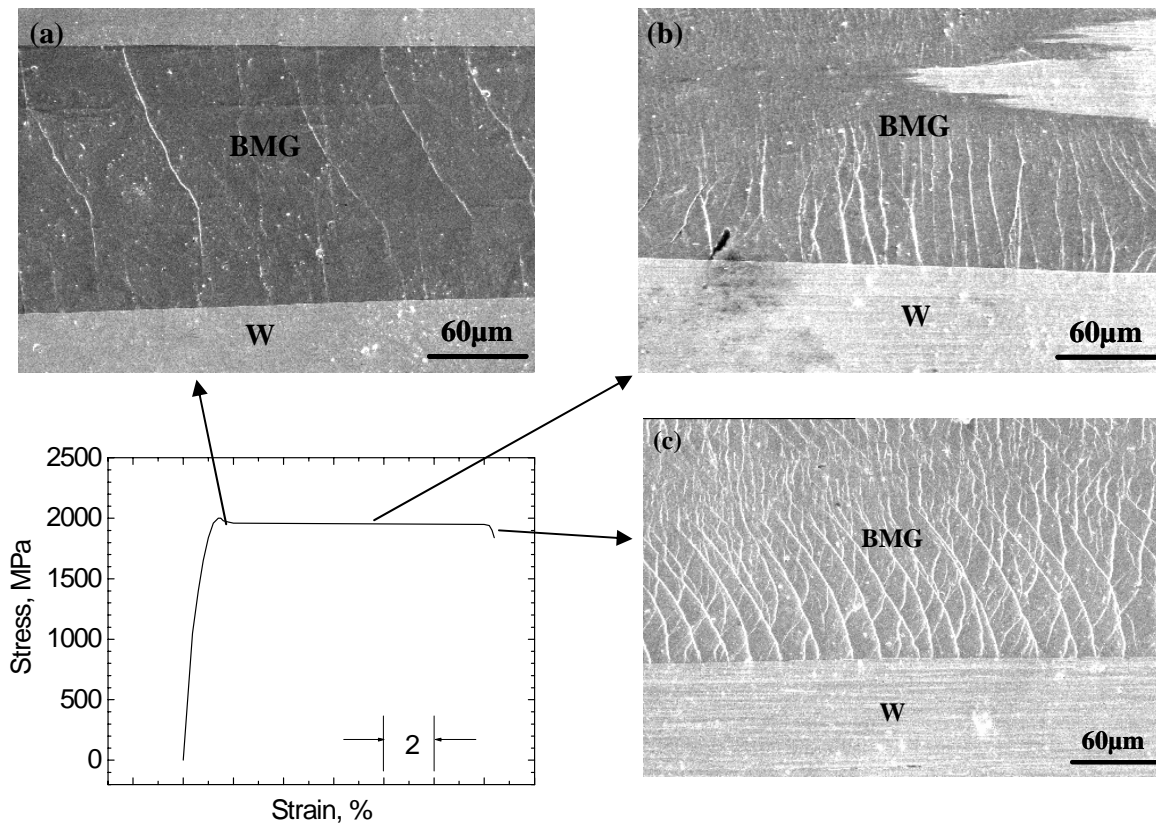


Figure 5.—SEM micrographs of shear band formation and propagation in BMG composite at different stages of compression test: (a) at the yielding point, (b) at the mid-point of plastic deformation, and (c) at the fracture point. The tungsten volume fraction is 60%.

are blocking the propagation of the single shear band. The multiple shear bands also propagate and then stop at the fiber/matrix interface. Further compression leads to more shear bands being generated in the BMG matrix to accommodate the strain (Figure 5b). Evidently the tungsten reinforcement provides large constraint to the shear bands propagation which prevents catastrophic failure of the materials. At the end, multiple shear bands are uniformly distributed in the entire volume of the glass matrix, as shown in Figure 5c. Choi-Yim et al [6] suggested that uniform shear band distributions occur when the initial slip displacement is small, otherwise localized multiple shear banding is expected. The constraints from the reinforcement also lead to branching of shear band (Figure 5c).

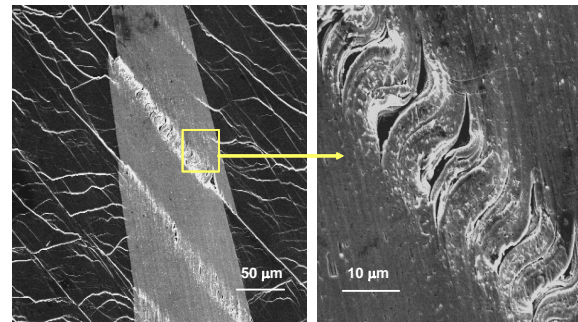


Figure 6.—Shear failure of tungsten fiber in BMG composite.

When the stress in the interface reaches to certain level, it will transfer to the fiber reinforcement. There are two mechanisms for

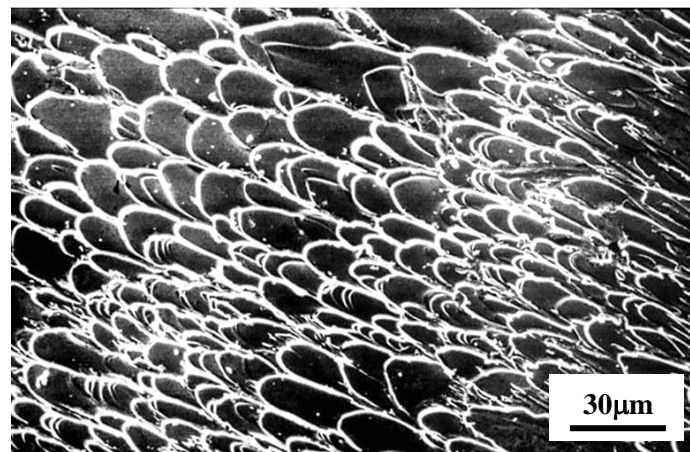
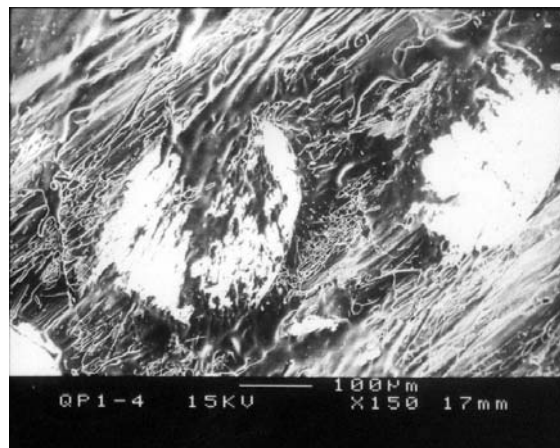
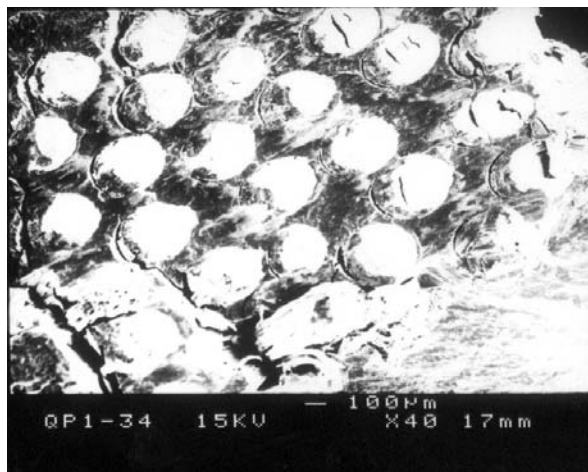


Figure 7.—SEM photograph of the compressive fracture surface of (a) BMG composite (40 vol. % tungsten), (b) is an enlarged image of (a), and (c) unreinforced BMG material.

the fracture energy dispersion. One is yielding and fracture of the reinforcement fiber, as shown in Figure 6. The multiple shear bands eventually lead to the shear fracture of the tungsten fiber. The other mechanism is fiber buckling and tilting. Conner et al has reported this mechanism in [4].

Figure 7 show the fracture surfaces of the monolithic BMG and the BMG composite under compression. Figure 7a & b are from 40% tungsten fiber reinforced composite. The tungsten fibers are tightly bound to the matrix because the thermal expansion coefficient for matrix ($8.5 \times 10^{-6}/^{\circ}\text{C}$) is almost twice that of the tungsten ($4.4 \times 10^{-6}/^{\circ}\text{C}$), resulting in high compressive radial stress at the fiber/matrix interface. In addition, the Poisson ratio of the matrix is higher than that of the reinforcement (0.35 versus 0.28), causing the matrix to squeeze the fibers even more tightly when the material is under compression. The fiber first yield, followed by axial shear cracking and also buckling. Figure 7c is the unreinforced BMG. It show a typical vein pattern, which is attributed to localized melting during shear band failure.

Conclusions

- A tungsten fiber reinforced beryllium-free Zr-based BMG composite was fabricated and characterized.
- Unlike the monolithic BMG, which usually fails on one shear band with very little plastic strain,. The BMG composite exhibits a large plastic deformation under compression. The plastic strain is up to 13%.
- The tungsten fiber reinforcement in the BMG composite provides constraint to the shear band propagation and promotes the formation of multiple shear bands. The stress will then transfer to the tungsten fiber, resulting in fiber shear fracture and buckling.

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References

1. Inoue, A. Stabilization of metallic supercooled liquid and bulk amorphous alloys. *Acta Mater* 2000, 48, 279-306.
2. Johnson, W. L. Bulk amorphous metal---an emerging engineering material. *JOM* 2002, March, 40-43.
3. Wang, W. H; Dong, C; Shek, C. H. Bulk metallic glasses. *Mater Sci. & Eng.* (2004), R44, 45-89.
4. Conner, R. D; Dandliker, R. B; Johnson, W. L. Mechanical properties of tungsten and steel fiber reinforced Zr-based metallic glass matrix composites. *Acta Mater* 1998, 46, 6089-6102.
5. Wang; H. Bulk metallic glass composites. *J. Mater. Sci. Technol.* 2005, 21 (suppl.1), 86-90.
6. Choi-Yim, H; Johnson, W. L. Bulk metallic glass matrix composites. *Appl. Phys. lett.* 1997, 71, 3808-3810.
7. Fu, H. M; Wang, H; Zhang, H. F; Hu, Z. Q. In-situ TiB-reinforced Cu-based bulk metallic glass composites. *Scripta Mater.* 2006, 54, 1961-1966.
8. Qiu, K. Q. PhD Thesis. Institute of Metal Research, Chinese Academy of Sciences, 2002.